

# **Time-Resolved X-ray Measurements of Polaron Dynamics of Charge-Ordered $\text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$**

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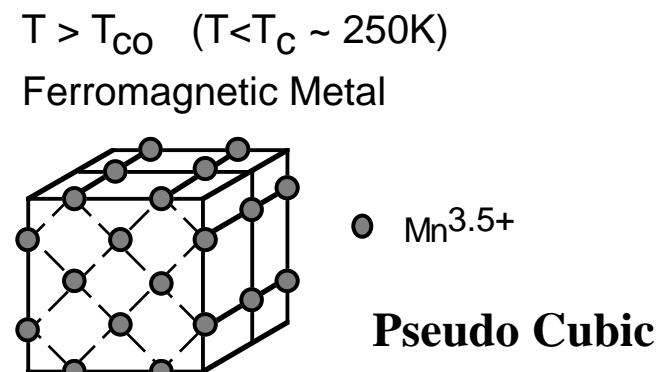
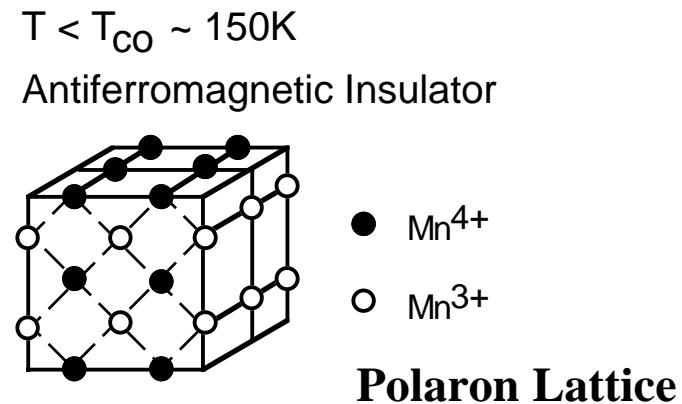
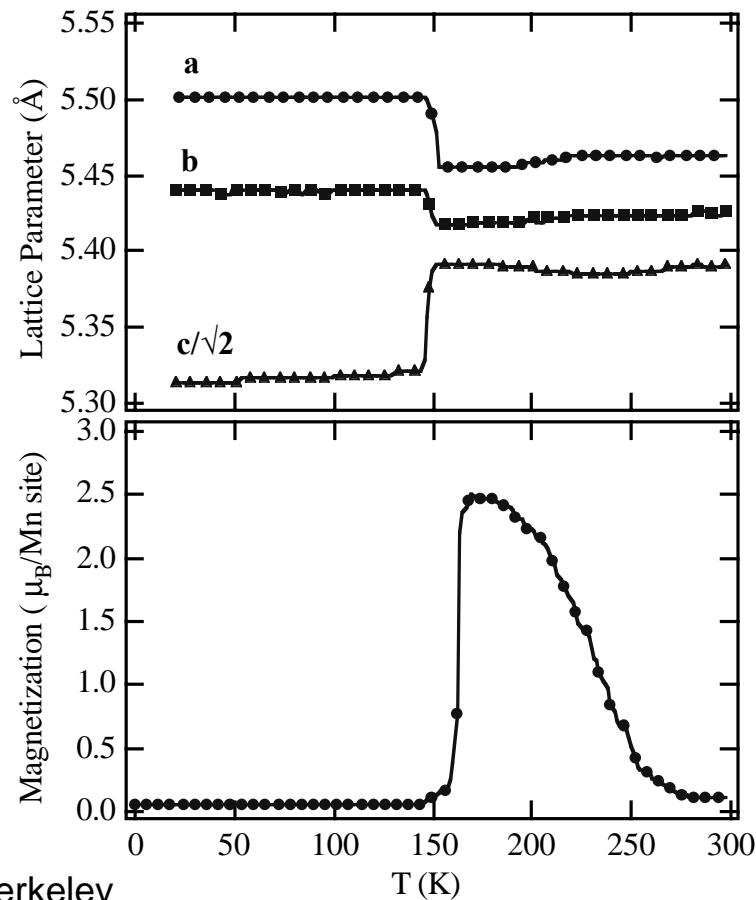
# **Time-resolved x-ray study of strongly-correlated materials**

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- **Dynamics of the strong charge-lattice-spin correlation in High-T<sub>c</sub> superconductors or CMR materials.**
- **Ultrafast x-ray techniques can obtain a complete picture of charge, spin, *and* lattice dynamics.**
  - » **Time-resolved X-ray diffraction -> charge-lattice interaction, the dynamics of orbital/charge ordering.**
  - » **Time-resolved EXAFS/NEXAFS -> local charge, lattice order.**
  - » **Time-resolved XMCD/NEXAFS -> charge-spin coupling.**

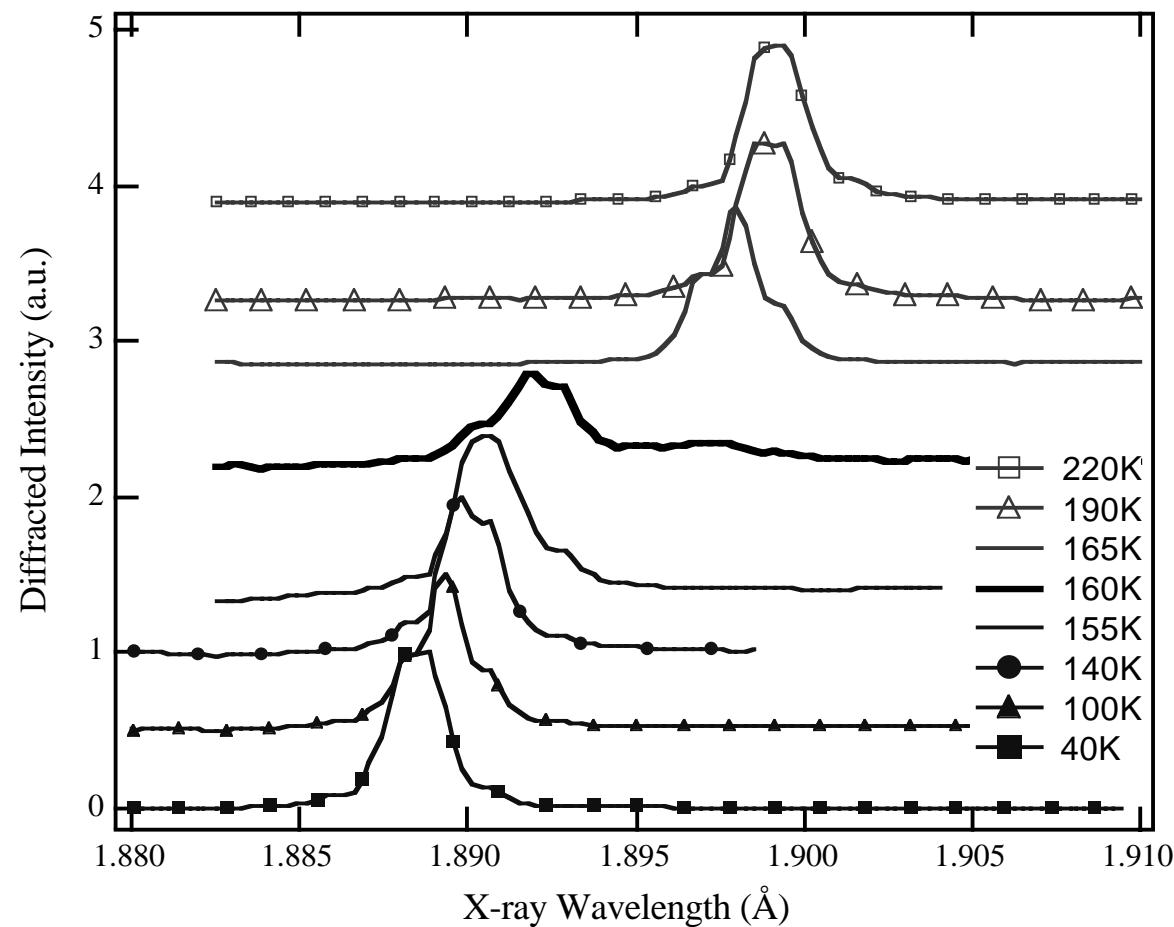
# $\text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ : a model system of strong charge-spin-lattice coupling

- Competition between FM and CO: mechanism for CMR?
- Data from Kuwahara *et al.*, PRB (1997)



# Rocking curve scans of (121) Bragg peak: first order phase transition at $T_{co}$

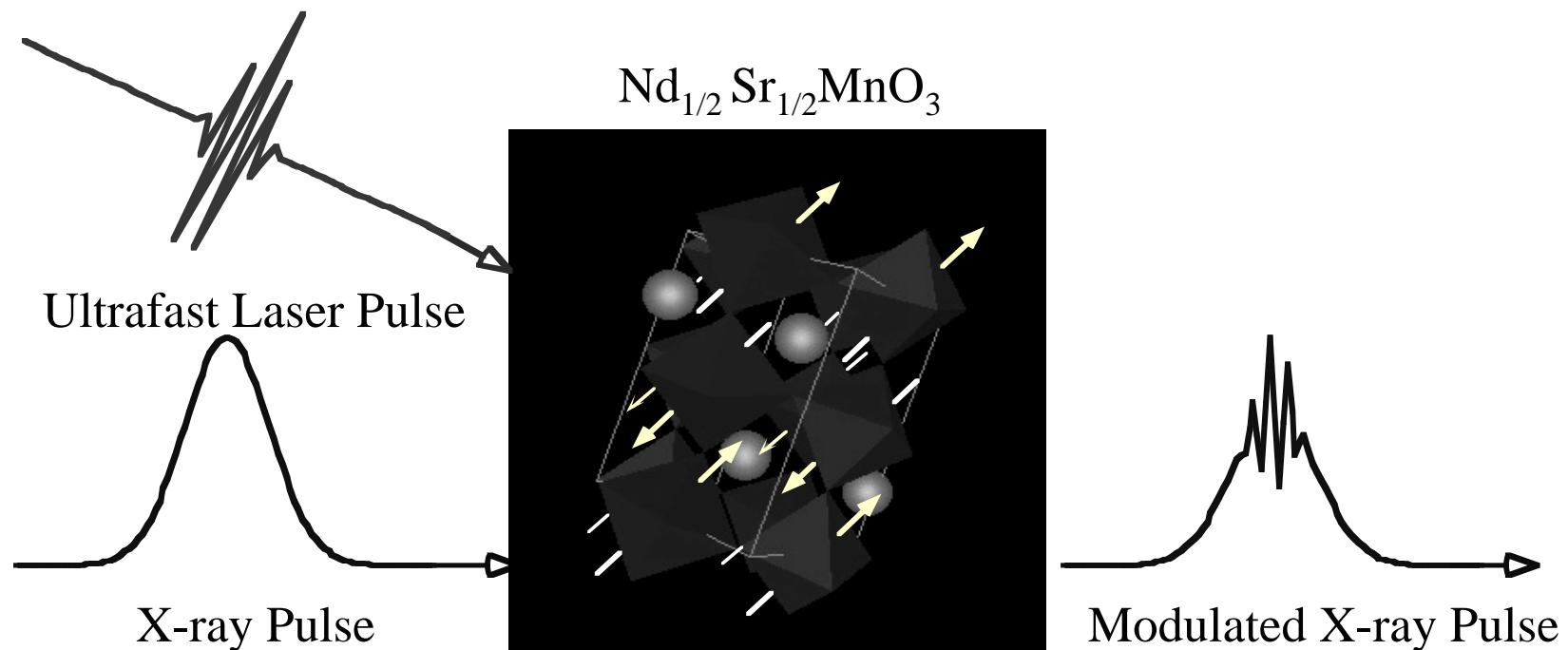
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Imma (121) = (101) of cubic perovskite

# Charge-transfer transitions and lattice dynamics in $\text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$

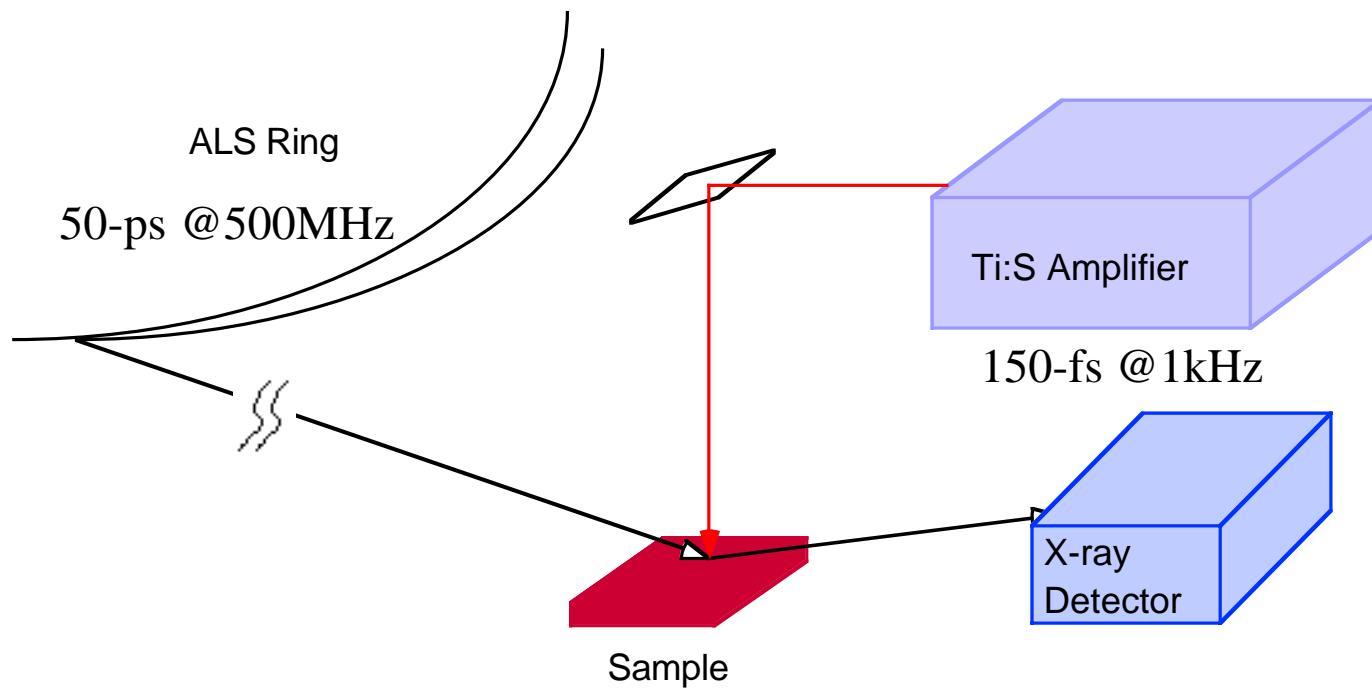
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- Ultrafast laser pulses induce charge-transfer transitions.
- Monitor the resultant lattice distortion via time-resolved x-ray diffraction.

# Setup: time-resolved x-ray diffraction of laser-perturbed solid

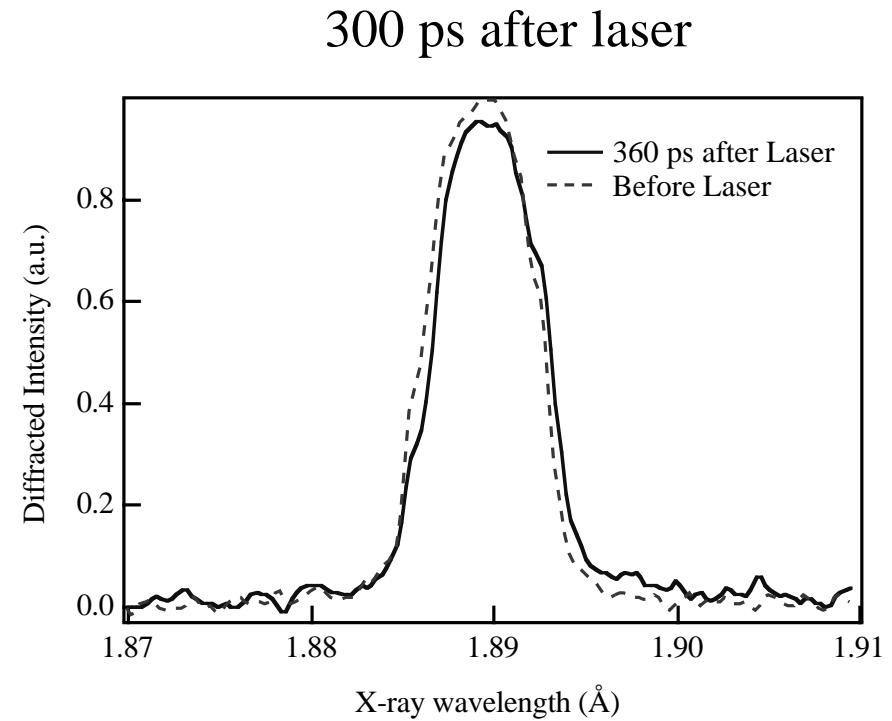
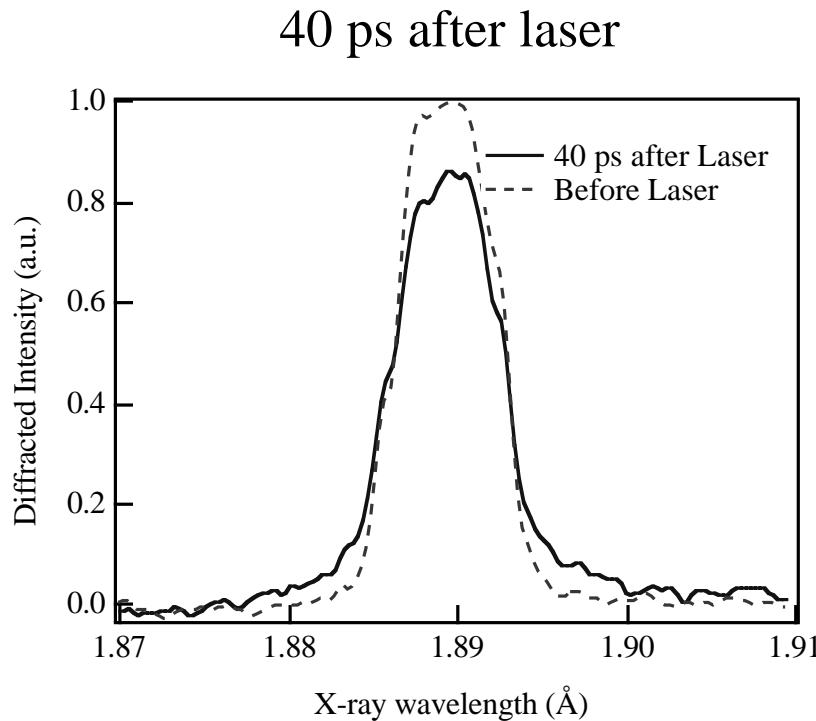
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# Ultrafast polaron dynamics

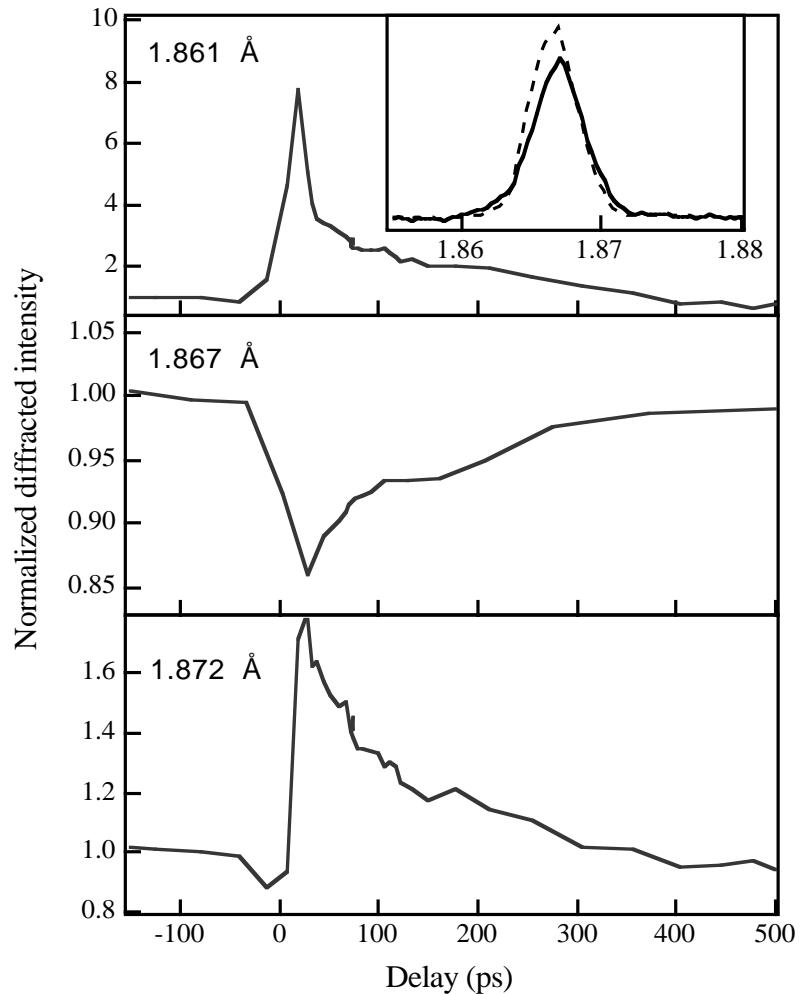
## Time-resolution = ALS pulse duration (~50 ps)

- Increase of x-ray scattering in the wings (within the time-resolution) and recovery in  $\sim 300$  ps : evidence of transient **polaron dynamics**. *cf.* S. Shimomura, et al. PRL (1999)



# Coherent laser-driven lattice strain

- The increase on both sides of the Bragg peak indicates the existence of compressive *and* expansive strain : not a thermal effect.
- Recovery time scale ~ time for the sound wave to traverse the x-ray probe depth : coherent strain wave.



# Rocking curve analysis

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- Diffraction amplitude: sum of the contribution from strained layer

$$X = i \sum_j^n a_j \exp[-i(A_j Y_j + \Phi_j)] \frac{\sin(A_j Y_j)}{Y_j} \quad \text{Diffraction Amp.}$$

$$a_j = \exp\left(-\frac{\gamma_o + \gamma_h}{2\gamma_o\gamma_h} \sum_{i=j+1}^n \mu_i t_i\right) \quad \text{X-ray absorption}$$

$$\Phi_j = 2 \sum_{i=j+1}^n A_i Y_i \quad \text{Propagation phase delay}$$

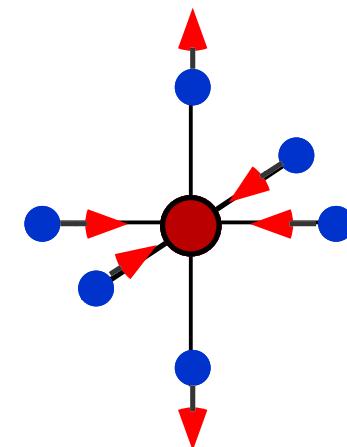
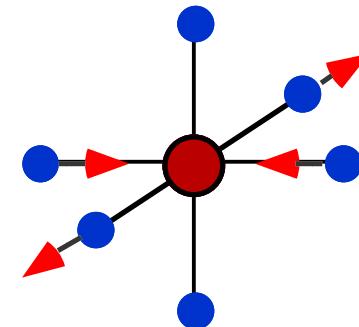
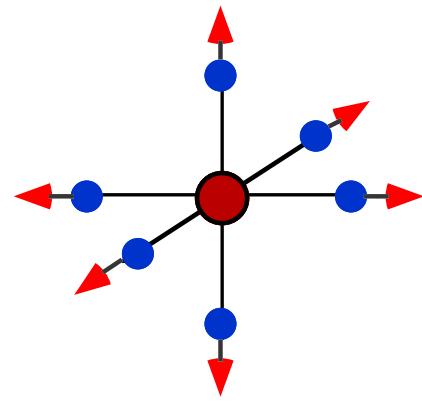
$$A_j Y_j \sim \sin 2\theta_B \Delta\theta$$

$$\Delta\theta \sim \Delta\theta + \tan\theta_B \varepsilon_L \text{ (long. strain)} + \varepsilon_T \text{ (trans. strain)}$$

# Charge-transfer and Jahn-Teller distortion

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- Impulsive charge transfers induce the Jahn-Teller lattice distortions around Mn ions.



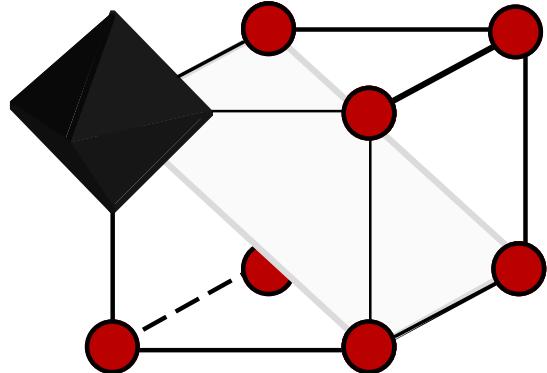
$$\sigma(Q_1) \sim \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

$$\sigma(Q_2) \sim \begin{pmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

$$\sigma(Q_3) \sim \begin{pmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 2 \end{pmatrix}$$

# Anisotropic strain waves induced by laser-driven Jahn-Teller distortion

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$$\sigma'(Q_1) \sim \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

Longitudinal expansion

$$\sigma'(Q_2) \sim \begin{pmatrix} -1 & 0 & -1 \\ 0 & 1 & 0 \\ -1 & 0 & -1 \end{pmatrix}$$

Longitudinal compression  
Transverse compression

$$\sigma'(Q_3) \sim \begin{pmatrix} 1 & 0 & -3 \\ 0 & -1 & 0 \\ -3 & 0 & 1 \end{pmatrix}$$

Longitudinal expansion  
Transverse compression

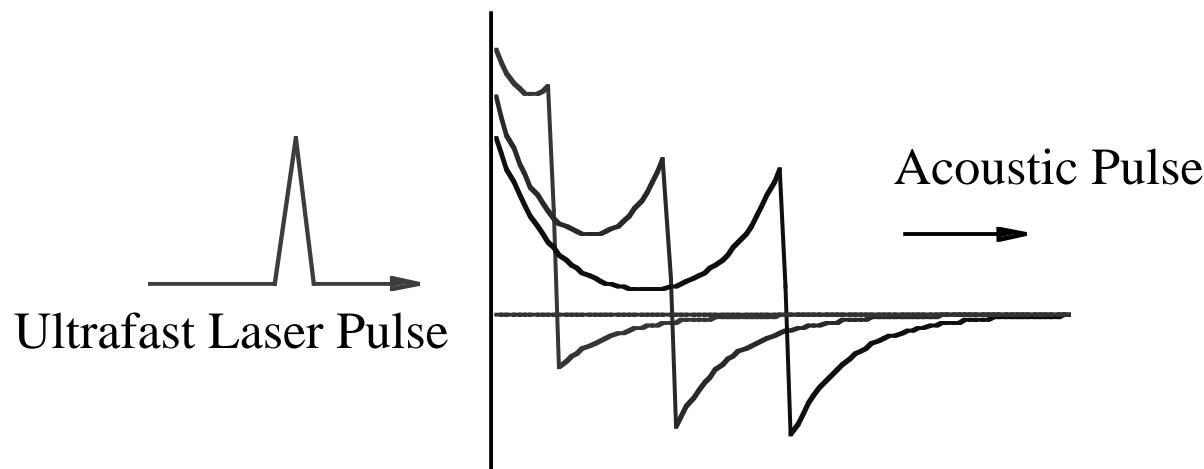


Mn ions

# Transient X-ray diffraction from polaron-induced strain waves

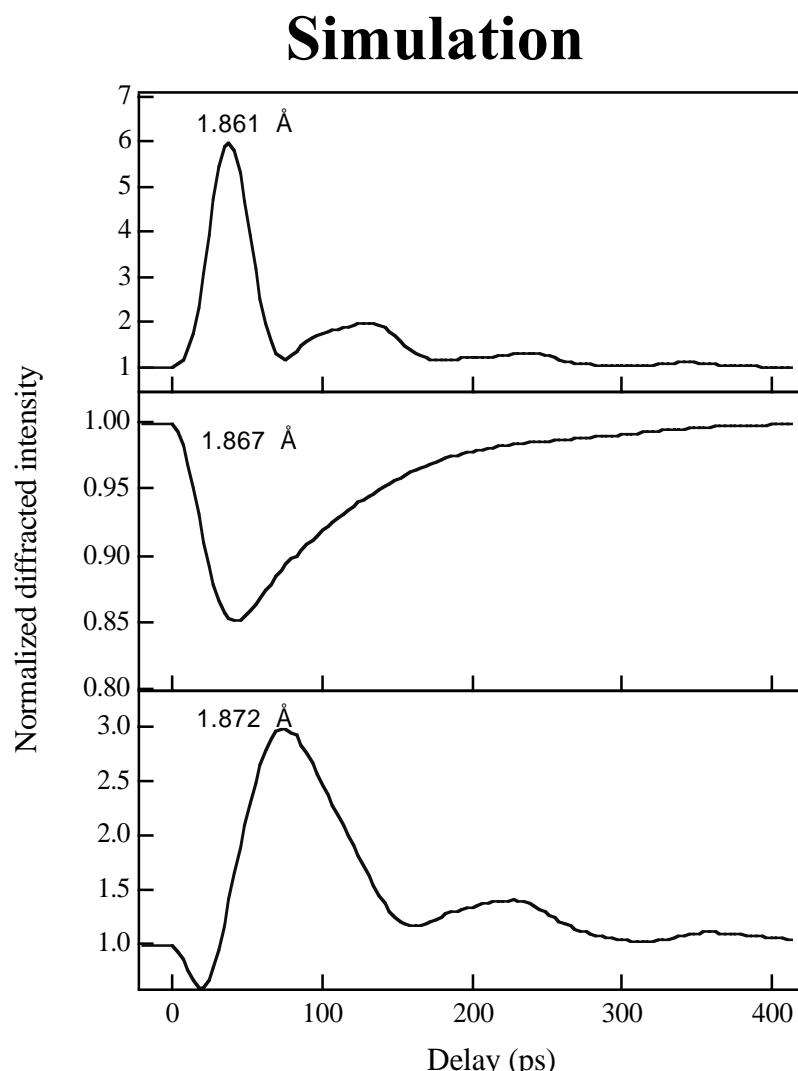
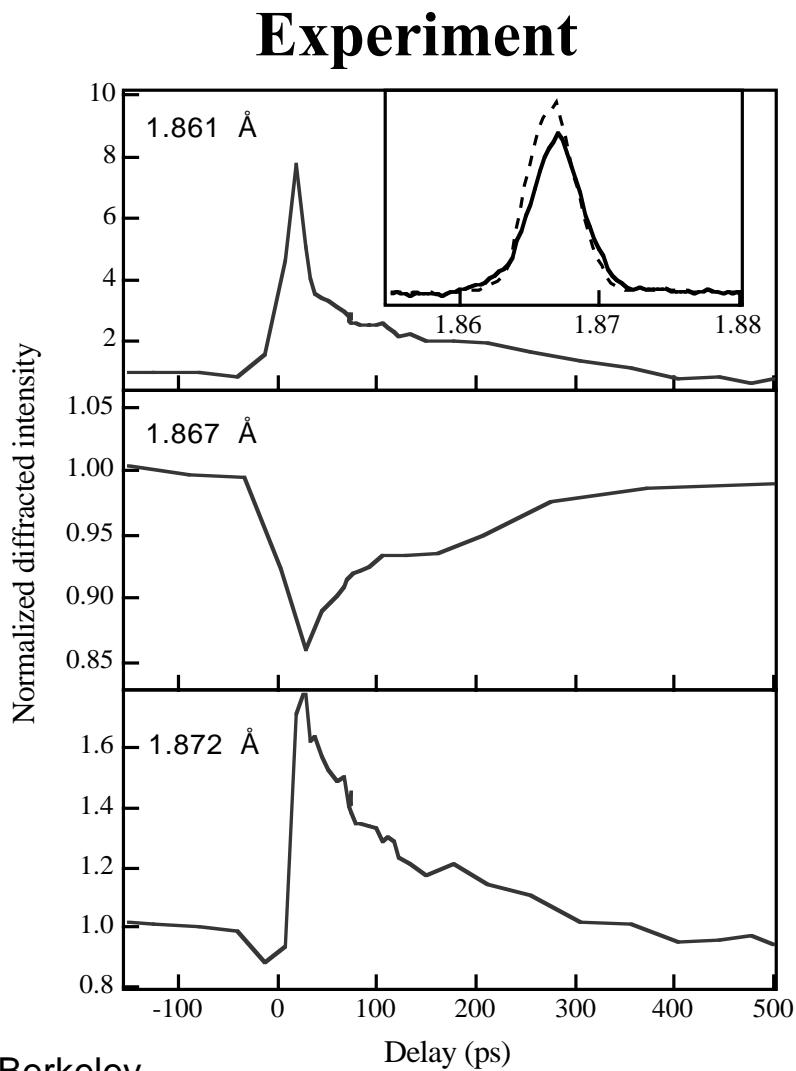
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- Impulsive excitation of JT polarons launches strain waves.



- $Q_1$  : Expansive longitudinal strain.
- $Q_2$  : Compressive longitudinal/transverse strain.
- $Q_3$  : Expansive longitudinal and compressive transverse strain.

# Experiment vs. Simulation with Jahn-Teller strain waves



# Results

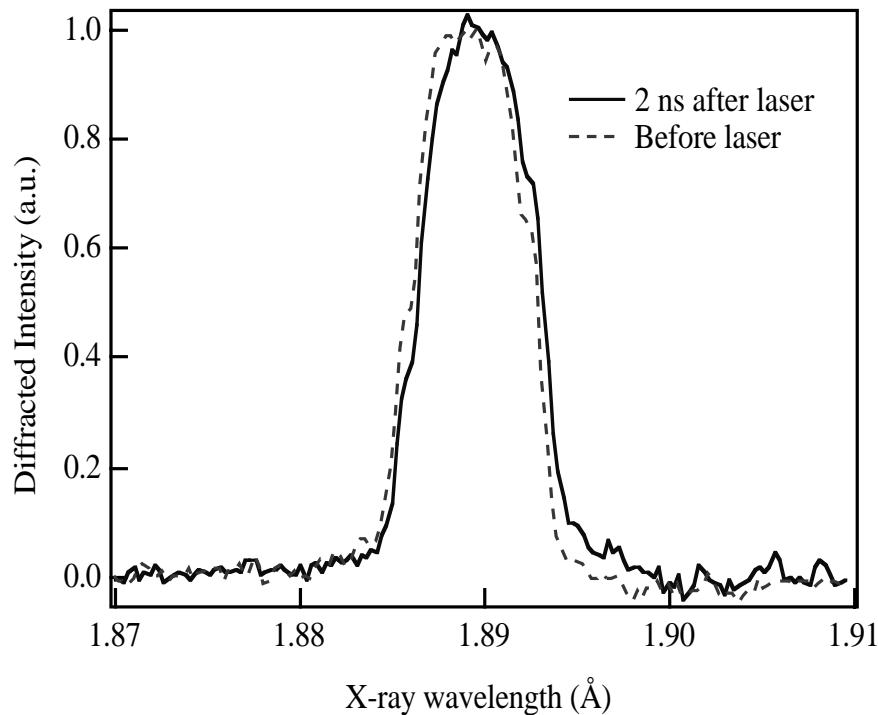
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- **Strong Coupling between charge transfer and lattice dynamics:**
  - » Charge-transfer induces  $Q_1$ ,  $Q_2$ , and  $Q_3$  distortions:  $Q_1$  and  $Q_3$  contribute to the signal.
  - » JT distortion energy  $\sim 1$  eV - not dependent on T.
  - » Charge-transfer anisotropy: indicative of an orbital ordering.
- **Polaron decays in  $\sim 50$  ps:**
  - » polaron  $\rightarrow$  ???  $\rightarrow$  thermal acoustic phonon.  
50 ps       $\sim 100$  ns

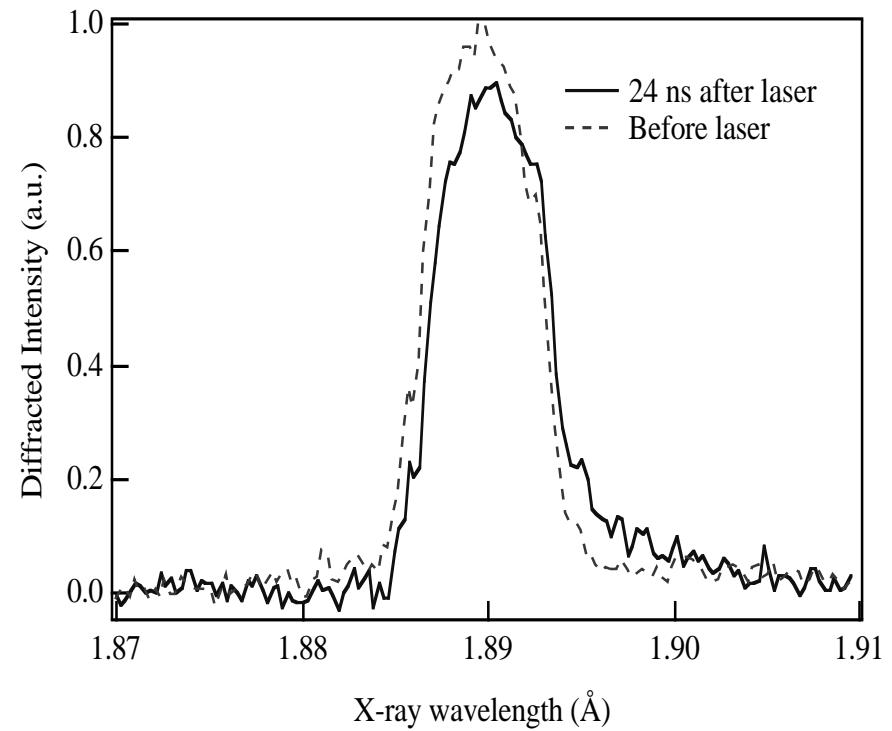
# Slow dynamics: Thermal strain develops ~ 100 ns after polaron decay

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**2 ns after laser**



**~20 ns after laser**



# **Conclusion and Future direction**

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- Ultrafast time-resolved x-ray diffraction clearly demonstrates the strong coupling between charge and lattice in CMR materials.
- The coupling strength is  $\sim 1$  eV independent of temperature.
- Polarons decay in  $\sim 50$  ps and subsequently into thermal acoustic phonons in  $\sim 100$  ns.
- Time-resolved soft x-ray MCD and NEXAFS for spin and charge-ordering dynamics.